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The direct effect of aerosols on solar radiation based on satellite observations, reanalysis datasets, and spectral aerosol optical properties from Global Aerosol Data Set (GADS)

N. Hatzianastassiou^{1,2}, C. Matsoukas^{2,3}, E. Drakakis^{2,5}, P. W. Stackhouse Jr.⁶,
P. Koepke⁷, A. Fotiadis^{2,4}, K. G. Pavlakis^{2,8}, and I. Vardavas^{2,4}

¹Laboratory of Meteorology, Department of Physics, University of Ioannina, 45110 Ioannina, Greece

²Foundation for Research and Technology-Hellas, Heraklion, Crete, Greece

³Department of Environment, University of the Aegean, Mytilene, Greece

⁴Department of Physics, University of Crete, Crete, Greece

⁵Department of Electrical Engineering, Technological Educational Institute of Crete, Heraklion, Greece

⁶Atmospheric Sciences, NASA Langley Research Center, Hampton, Virginia, USA

⁷Meteorological Institute, University of Munich, Munich, Germany

⁸Department of General Applied Science, Technological Educational Institute of Crete, Heraklion, Greece

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Correspondence to: N. Hatzianastassiou (nhatzian@cc.uoi.gr)

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Abstract

A global estimate of the seasonal direct radiative effect (DRE) of natural plus anthropogenic aerosols on solar radiation under all-sky conditions is obtained by combining satellite measurements and reanalysis data with a spectral radiative transfer model.

5 The estimates are obtained with detailed spectral model computations separating the ultraviolet (UV), visible and near-infrared wavelengths. The global distribution of spectral aerosol optical properties was taken from the Global Aerosol Data Set (GADS) whereas data for clouds, water vapour, ozone, carbon dioxide, methane and surface albedo were taken from various satellite and reanalysis datasets. Using these aerosol
10 properties and other related variables, we generate climatological (for the 12-year period 1984–1995) monthly mean aerosol DREs. The global annual mean DRE on the outgoing SW radiation at the top of atmosphere (TOA, ΔF_{TOA}) is 1.62 Wm^{-2} (with a range of -10 to 15 Wm^{-2} , positive values corresponding to planetary cooling), the effect on the atmospheric absorption of SW radiation (ΔF_{atmab}) is 1.6 Wm^{-2} (values up
15 to 35 Wm^{-2} , corresponding to atmospheric warming), and the effect on the surface downward and absorbed SW radiation (ΔF_{surf} , and $\Delta F_{\text{surfnet}}$, respectively) is -3.93 and -3.22 Wm^{-2} (values up to -45 and -35 Wm^{-2} , respectively, corresponding to surface cooling.) According to our results, aerosols decrease/increase the planetary albedo by -3 to 13% at the local scale, whereas on planetary scale the result is an increase of
20 1.5% . Aerosols can warm locally the atmosphere by up to 0.98 K day^{-1} , whereas they can cool the Earth's surface by up to -2.9 K day^{-1} . Both these effects, which can significantly modify atmospheric dynamics and the hydrological cycle, can produce significant planetary cooling on a regional scale, although planetary warming can arise over highly reflecting surfaces. The aerosol DRE at the Earth's surface compared to TOA
25 can be up to 15 times larger at the local scale. The largest aerosol DRE takes place in the northern hemisphere both at the surface and the atmosphere, arising mainly at ultraviolet and visible wavelengths.

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1 Introduction

Atmospheric aerosols, both natural and anthropogenic, can cause climate change through their direct, indirect and semi-direct effects on the radiative energy budget of the Earth-atmosphere system. The sum of these effects, on a global average, is likely to be comparable in magnitude to the radiative forcing of about 2.4 Wm^{-2} by anthropogenic greenhouse gases (IPCC, 2001). However, the quantification of the aerosol effects is more complex than the quantification of radiative forcing by greenhouse gases because aerosol mass and particle number concentrations are highly variable in space and time, due to their much shorter atmospheric lifetime compared with the important greenhouse gases. Aerosol properties are difficult to measure without instrumental offsets or bias, making it difficult to estimate the aerosol radiative effects. Therefore, there is large uncertainty regarding the aerosol overall radiative forcing (Remer and Kaufman, 2006; Granger Morgan et al., 2006; Yu et al., 2006). Better estimates of the aerosol radiative effects on a planetary scale are required to reduce the uncertainties.

The present study focuses on the direct radiative effect (DRE) of aerosols, which is the overall effect of natural plus anthropogenic aerosols on the radiative energy budget, and it is different to the direct climate forcing (DCF) which represents the corresponding effect of anthropogenic aerosols only. Although the present consensus is that the DRE is better known than the indirect and semi-direct effects, there is still a large degree of uncertainty. Our study deals with the direct effect of aerosols on the shortwave (SW) radiation budget, since the main DRE lies at these wavelengths, being much smaller in the longwave (thermal infrared). Even for dust, the thermal infrared radiative effects are about 10% those for the shortwave radiation (Tanré et al., 2003).

The assessments of aerosol DRE fall into three broad categories: (i) measurement-based (e.g. Bellouin et al., 2005; Loeb and Manalo-Smith, 2005; Zhang et al., 2005; Remer and Kaufman, 2006; Christopher et al., 2006), (ii) model-based (e.g. Jacobson, 2001; Chin et al., 2002; Liao et al., 2004; Koch and Hansen, 2005; Reddy et al., 2005; Takemura et al., 2005), and (iii) measurement-model integrated (e.g. Chu et

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al., 2002; Yu et al., 2004; Chung et al., 2005). Measurements involve in-situ, satellite and suborbital remote sensing data. Yu et al. (2006) provide a review of the above assessments. Each method has advantages and drawbacks. For example, there is a wide range of discrepancy in model results because of the many inherent assumptions involved in modelling the aerosol effect on climate (Remer and Kaufman, 2006), so there are difficulties in reproducing correctly satellite observations (Bellouin et al., 2005; Zhao et al., 2005). On the other hand, although many problems related to satellite observations of aerosols have been resolved by more sophisticated instruments, there are still problems as shown by discrepancies between different satellite products (Zhao et al., 2005). Beyond this, the satellite-based method (e.g. Christopher and Zhang, 2002) provides estimates of aerosol forcing only at the top of atmosphere (TOA), but not within the atmosphere and at the Earth's surface, where the effect of aerosols is largest.

The present study takes an alternate approach using quality available global observational data and detailed spectral radiative transfer flux calculations performed with a radiative transfer model. The adopted technique relies on spectral aerosol optical properties from the Global Aerosol Data Set (GADS, Koepke et al., 1997) and satellite observations for the key surface and atmospheric parameters such as clouds, water vapour, surface albedo and ozone taken from comprehensive global climatological databases (NASA Langley Research Center data set, International Satellite Cloud Climatology Project, ISCCP, Goddard Earth Observing System, GEOS, version 1 reanalysis, provided by the Data Assimilation Office, DAO, of NASA's Goddard Space Flight Center, GSFC). This study incorporates realistic surface and atmospheric conditions, especially those of clouds, to reduce some of the major sources of error in estimates of the direct forcing/effect (Podgorny and Ramanathan, 2001; Chung et al., 2005). In addition, an important improvement in this study, compared to other studies, is that the aerosol DRE is computed at a very high spectral resolution, while other radiation transfer, general circulation (GCM) and chemical transport (CTM) models, usually include a few spectral bands in the whole SW range. This can be critical, since inadequate

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treatment of spectrally resolved aerosol properties can result in modified aerosol forcings of up to 16% on a global scale, and 100% locally (Hatzianastassiou et al., 2004b). Recently, Redemann et al. (2006) have shown that ratios of aerosol radiative forcing at 400 and 1600 nm can be as high as a factor of 10, depending on the aerosol type. Our DRE computations refer to the whole solar spectrum (0.2–10 μm), and they are performed at 117 wavelengths ranging from 0.2 to 1.0 μm , and 10 spectral intervals in the range 1.0–10.0 μm (note that our study deals only with solar radiation). The inclusion of near-IR has been shown (e.g. Bush and Valero, 2003; Nishizawa et al., 2004; Zhou et al., 2005; Hatzianastassiou et al., 2006) to be essential for accurately computing the aerosol radiative effects. We have used a detailed spectral radiative transfer model to match, as best possible, the detailed spectral resolution of aerosol optical properties (extinction optical thickness, AOT, single scattering albedo, ω_{aer} , and asymmetry parameter, g_{aer}) from GADS, given at 40 wavelengths within the range 0.2 to 10.0 μm , using interpolation and extrapolation when necessary.

Following this approach, we produced global DRE monthly climatologies for the 12-year period 1984–1995 for both clear and all-sky conditions, for winter and summer. The DRE is computed at TOA, in the atmosphere and at the Earth's surface. All computations were performed at a geographical cell level of $1^\circ \times 1^\circ$ latitude-longitude on a daily basis, and subsequently averaged and presented as monthly means. Our results provide a realistic climatological assessment of aerosol SW DRE.

This study complements the works by Hatzianastassiou et al. (2004a, 2006). In Hatzianastassiou et al. (2004a) the global distribution of aerosol DRE in the ultraviolet and visible wavelengths was computed for clear-sky conditions, for the 10-year period 1984–1994. Recently, Hatzianastassiou et al. (2006) calculated the aerosol DRE in the solar near-infrared (IR) for both clear- and all-sky conditions for the period 1984–1995. The present study is an integration of the previous two studies, providing estimates of clear- and all-sky DREs for the total SW range for the period 1984–1995.

The methodology, and descriptions of the radiative transfer model and the climatological input data were given in detail in Hatzianastassiou et al. (2004a, 2006), so only

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a very brief description of these will be given in Sect. 2. More emphasis will be given to the results (Sect. 3), before the conclusions (Sect. 4).

2 Model and input data

The radiative transfer model and the various assimilated input data are briefly described in this section. For more details the reader is referred to the relevant studies by Hatzianastassiou et al. (2004 a,b) and Hatzianastassiou et al. (2006).

2.1 The radiative transfer model

The deterministic spectral radiative transfer model used here was developed from a radiative-convective model (Vardavas and Carver, 1984). The incoming solar irradiance at TOA is computed for the spectral interval 0.2–10.0 μm , based on the spectral profile of Thekaekara and Drummond (1971), using a solar constant $S_0=1367\text{ Wm}^{-2}$ (e.g. Willson, 1997) corrected for the Earth's elliptical orbit. The computations were performed for each 1-degree latitude-longitude cell for 117 wavelengths in the range 0.2–1.0 μm and ten spectral intervals in the range 1.0–10 μm . For each wavelength and spectral interval, a set of monochromatic radiative flux transfer equations is solved for an absorbing/multiple-scattering atmosphere using the Delta-Eddington method of Joseph et al. (1976) which is an extension of the Eddington method described in Shettle and Weinman (1970). At ultraviolet–visible wavelengths we include ozone absorption and Rayleigh scattering by air. For the near-IR wavelengths we include absorption by water vapour, carbon dioxide and methane. We include scattering and absorption by clouds (low, middle and high) and aerosols, and reflection from the Earth's surface.

The SW aerosol DRE (denoted henceforth as ΔF), or more precisely the “aerosol flux change”, is the effect of aerosols on the SW radiation budget at TOA, at the Earth's surface, or within the atmosphere, and it is given by

$$\Delta F = F - F_{\text{no-aerosol}} \quad (1)$$

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where F and $F_{\text{no-aerosol}}$ are the SW radiative fluxes with and without aerosols. The DRE components ΔF_{TOA} , ΔF_{atmab} , ΔF_{surf} , and $\Delta F_{\text{surfnet}}$, represent the effect of aerosols on the outgoing radiation at TOA, the radiation absorbed within the atmosphere, the downward radiation at the Earth's surface, and the net downward (or absorbed) radiation at the surface.

2.2 GADS aerosol data

The aerosol radiative properties, AOT, ω_{aer} , and g_{aer} , were taken from GADS (Koepke et al., 1997), which provides aerosol particle properties averaged over space and time, used in a number of climate model studies (e.g. King et al., 1999; Chin et al., 2002; Morcrette, 2002; Treffeisen et al., 2005). Detailed discussions of GADS aerosol properties were given in Hatzianastassiou et al. (2004a, 2006). In GADS, the tropospheric aerosol particles are described by 10 main aerosol components, which are representative for the atmosphere, resulting from aerosol emission, formation, and removal processes within the atmosphere, so that they exist as mixture of different substances, both external and internal. Global distributions of GADS aerosol properties are given as climatologically averaged values both for the periods December through February (northern hemisphere winter) and June through August (northern hemisphere summer) on a $5^\circ \times 5^\circ$ latitude-longitude resolution. However, to match the spatial resolution of the climatological parameters, especially that of relative humidity to which the aerosol properties are sensitive, the original GADS aerosol optical properties were up-scaled to $1^\circ \times 1^\circ$ latitude-longitude resolution, as explained in Hatzianastassiou et al. (2006). The aerosol properties originally taken from GADS, were re-computed for actual relative humidity values for the aerosol layer in order to compute realistically the aerosol DREs.

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2.3 Surface and atmospheric data

The water vapour and temperature data were taken from the GEOS-1 reanalysis product (6-hourly), averaged to a daily temporal resolution. Atmospheric temperature and specific humidity profiles for the years 1984 through to 1995 were used to compute the mean relative humidity of the aerosol layer. Cloud data on a $1^\circ \times 1^\circ$ resolution were taken from the NASA-Langley data set based on GEWEX ISCCP DX cloud climatologies. The mean daily $1^\circ \times 1^\circ$ NASA-Langley cloud data were compiled by processing the 3-hourly ISCCP-DX pixel-level data, which contain radiance and cloud retrieval information from geosynchronous and polar orbiting satellites sampled to a nominal resolution of 30 km. All 30 km DX pixels within a grid cell are averaged analogously to the methods of ISCCP (e.g. Rossow et al., 1996) to produce gridded radiance and cloud products. The NASA Langley $1^\circ \times 1^\circ$ cloud data include: cloud amount, cloud-top pressure, cloud-top temperature, liquid water path, and optical depth for total clouds. They also provide cloud amount and cloud-top temperature for low-, mid-, and high-level clouds, as well as cloud amount, cloud-top temperature, cloud optical depth, and cloud albedo separately for ice and liquid water phase clouds. The surface reflection was computed by using surface-type cover fractions on a mean daily and 1-degree geographical cell resolution for the years 1984–1995. Ice/snow cover data were taken from ISCCP, while surface type classification maps were obtained from other high resolution data sets (Stackhouse et al. 2002). A complete topography scheme is included in the model, which uses the NASA DAO GEOS-1 surface pressure, gridded on $1^\circ \times 1^\circ$ cells.

3 Shortwave aerosol direct radiative effect computations

The model total SW mean monthly (winter and summer) aerosol DREs for each cell (1° longitude-latitude) are given at TOA, in the atmosphere and at the surface. At TOA, the change in planetary albedo (ΔR_p) due to the presence of aerosols is also given.

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3.1 Aerosol direct radiative effect at the top of atmosphere(ΔF_{TOA})

The average (1984–1995) change of outgoing SW radiation (OSR) at TOA due to aerosols (aerosol direct radiative effect ΔF_{TOA}), for all-sky conditions, is given in Fig. 1. According to Eq. (1), positive values indicate increased OSR, i.e. solar radiative cooling effect of the Earth-atmosphere system. In contrast, negative ΔF_{TOA} values indicate decreased OSR due to aerosols, or a warming radiative effect. Given that the incoming SW radiation remains unchanged, the change of planetary albedo (ΔR_p) is directly dependent on ΔF_{TOA} . Thus, positive and negative values of ΔR_p in Fig. 2 correspond to increased and decreased planetary albedo, respectively, due to aerosols (expressed in absolute percentage values).

In general, natural plus anthropogenic aerosols modify the reflected SW radiation by the Earth-atmosphere system by about -10 to 15 Wm^{-2} (or -3 to 13% in terms of ΔR_p , Fig. 2). Over most of the globe the change is smaller than 5 Wm^{-2} (or 2%). Aerosols mostly increase the OSR (positive values of ΔF_{TOA} and ΔR_p) producing thus planetary cooling. However, they also produce a warming of the Earth-atmosphere system by decreasing the outgoing SW radiation at TOA by up to 10 Wm^{-2} (or -3% , Fig. 2). This occurs over high-latitude regions with strong surface albedo, such as Greenland in July and Siberia in January, as well as over continental areas (e.g. Sahara, Arabian peninsula) with large surface albedo and significant loads of absorbing aerosols. The sign of DRE at TOA is determined by ω_{aer} , AOT and surface albedo, R_g . Therefore, combinations of these parameters can lead to a changing sign of ΔF_{TOA} and ΔR_p with time over the same location (e.g. planetary warming over Siberia in winter against planetary cooling in summer), and also to a different sign of ΔF_{TOA} and ΔR_p over adjacent areas (e.g. planetary cooling over sub-Sahel and along the Atlantic Ocean tropical zone extending from northern Africa through central America, against planetary warming over highly reflecting Saharan desert areas). The role of ω_{aer} for ΔF_{TOA} is demonstrated by the fact that our study gives planetary warming over Sahara and Arabia (Figs. 1 and 2), whilst a cooling was found by Reddy et al. (2005) using $\omega_{\text{aer}} > 0.95$.

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Although planetary warming over these regions is suggested by other results (e.g. Yu et al. 2004), there is evidence (e.g. Kaufman et al. 2001; Dubovik et al. 2002; Cattrall et al. 2003) that dust absorption could be much weaker. To assess a possible overestimation of dust absorption by GADS, we performed a sensitivity analysis that showed that a 6% larger ω_{aer} produces a ΔF_{TOA} with opposite sign, i.e. planetary cooling over Sahara and Arabia, in agreement with Reddy et al. (2005).

Planetary warming produced by aerosols in high-latitude and polar regions, such as Greenland, are of much interest because of its relation to climate change issues (melting of ice, e.g. Rignot and Kanagaratnam, 2006). Negative DRE values of similar or even stronger magnitude are also expected at least over the Arctic in January, because of the presence of Arctic haze (e.g. Hu et al., 2005; Yamanouchi et al., 2005) involving long-range transport of anthropogenic pollution from industrialized areas in Europe, North America and Asia. Such features do not appear in Figs. 1 and 2, however, because they correspond to areas with missing data.

The largest values of ΔF_{TOA} occur in tropical and subtropical latitudes, and especially over continental desert areas (e.g. Sahara, Arabian peninsula, Taklimakan and Gobi in Asia, and central Australia), over oceanic areas over which are transported dust and smoke from biomass burning (e.g. off the coast of west Africa in the tropical Atlantic Ocean, Arabian Sea) and over remote oceanic areas with intense production of sea-spray (storm-track zone of the Southern Hemisphere), especially in January. The features shown in Fig. 1 are in agreement with those found from satellite-based studies (e.g. Loeb and Manalo-Smith, 2005; Yu et al., 2006), though some other features such as those associated with dust transport from Asia across the Pacific Ocean, are not well captured by GADS properties, perhaps due to the different periods considered (i.e. 1984-1995 here, against after 2000 in satellite-based studies). The magnitude of ΔF_{TOA} and ΔR_p for a specific place on the globe is determined by AOT, cloud cover and surface albedo, apart from the incoming solar flux. Thus, large values, up to 9 Wm^{-2} , appear in regions with small cloudiness and significant aerosol loads, e.g. sub-Sahel, Gobi and Australian deserts, as well as Middle-East and India in January. In July,

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large positive values (up to 15 Wm^{-2}) exist over limited maritime areas such as the eastern Mediterranean basin and the Red Sea, and secondarily over continental areas such as Australia. Note that highly populated and industrialized urban areas such as Europe and North America are found to have relatively smaller values of ΔF_{TOA} despite relatively large AOT values, especially in the visible wavelengths. We note that small values of aerosol DRE at TOA do not exclude important aerosol effects taking place within the Earth-atmosphere system, since the effect at TOA is the sum of the effects of aerosols in the atmosphere and at the surface. The aerosol effects on the radiation budgets of the atmosphere and surface are examined separately, in the following two subsections.

Overall, the large modifications of OSR produced by aerosols on a climatological basis, equivalent to changes in planetary albedo of $\Delta R_p = \pm 1\text{--}2\%$ at the geographical cell level, can have a strong climatic impact. Aerosol modifications to spatial gradients in OSR can affect general circulation patterns in the atmosphere and hence climate, as shown by GCM studies (Kristjánsson et al., 2005; Lau et al., 2006).

3.2 Aerosol direct radiative effect in the atmosphere (ΔF_{atmab})

The aerosol DRE on the atmospheric absorption of SW radiation under all-sky conditions, ΔF_{atmab} , is very important and can be as large as 35 Wm^{-2} (Fig. 3). Nevertheless, over the oceans it is generally $\Delta F_{\text{atmab}} < 3 \text{ Wm}^{-2}$, whereas over most continental areas it is $\Delta F_{\text{atmab}} < 15 \text{ Wm}^{-2}$. The largest values of ΔF_{atmab} ($15\text{--}35 \text{ Wm}^{-2}$) are found over areas characterized by significant amounts of absorbing aerosols (such as mineral-dust or soot), especially over highly reflecting surfaces (surface albedo > 0.3) in tropical and subtropical latitudes, but large values are also found over the Middle-East, South and South-East Asia, Europe, USA, South America, South Africa, and Australia. Large values of ΔF_{atmab} are found over oceans only for transported aerosols of continental origin. This occurs across the tropical Atlantic due to long-range transport of desert dust at higher atmospheric levels (e.g. Formenti et al., 2003; Moulin and Chi-

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apello, 2004) by the “harmattan” trade winds during the dry season (this transport is shifted northwards in July due to the changed atmospheric circulation, Azores anticyclone and thermal low of Pakistan), as well as over the Yellow and Japan Seas due to Asian dust exported from the Gobi desert (e.g. Sun et al., 2001; Liu et al., 2003). The strong increase in atmospheric absorption of solar radiation is either associated with large mass concentrations of absorbing mineral aerosol components or with significant (but smaller) concentrations of strongly absorbing soot or water soluble and insoluble components, especially in the presence of large amounts of solar radiation and over highly reflecting surfaces underneath. The enhancement of absorbed solar radiation is converted into heat. The resulting atmospheric heating rate can be calculated from the First Law of Thermodynamics and hydrostatic equilibrium

$$\frac{\partial T}{\partial t} = \frac{g}{c_p} \frac{\Delta F}{\Delta P} \quad (2)$$

where $\partial T/\partial t$ is the heating rate (K day^{-1}), g is the acceleration due to gravity, c_p the specific heat capacity of air at constant air pressure ($\sim 1006 \text{ J Kg}^{-1} \text{ K}^{-1}$) and P is the atmospheric pressure, respectively. The estimated ΔF_{atmab} translates into an atmospheric heating rate of 0.98 K day^{-1} for $\Delta F_{\text{atmab}} = 35 \text{ Wm}^{-2}$ for $\Delta P = 300 \text{ mb}$ (mid-latitude pressure width of troposphere), whereas the atmospheric heating rate over oceans is $< 0.14 \text{ K day}^{-1}$ (for $\Delta F_{\text{atmab}} < 5 \text{ Wm}^{-2}$).

3.3 Aerosol direct radiative effect at surface ($\Delta F_{\text{surfnet}}$)

Through scattering and absorption, together natural and anthropogenic aerosols decrease drastically the downwelling and absorbed solar radiation at the Earth’s surface by up to 45 and 35 Wm^{-2} , respectively. As shown in Fig. 4, large values of $\Delta F_{\text{surfnet}}$ are found over continental areas, especially over deserts and polluted regions of the world. Thus, the absorbed SW radiation by the surface is decreased by $20\text{--}35 \text{ Wm}^{-2}$ in the Sahara and Sahel, by $10\text{--}20 \text{ Wm}^{-2}$ in the sub-Sahel, Arabian peninsula and Middle-East, and by $5\text{--}15 \text{ Wm}^{-2}$ in Europe, India, eastern China and USA. The aerosol optical

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thickness, AOT, is the determinant parameter for $\Delta F_{\text{surfnet}}$. Smaller $\Delta F_{\text{surfnet}}$ values ($< 5\text{--}7\text{ W m}^{-2}$) are found over oceans, due to the optically thin aerosol layers there. Only in oceanic areas close to land and areas where long-range transport of aerosols takes place, the values of $\Delta F_{\text{surfnet}}$ become as large as $10\text{--}15\text{ W m}^{-2}$. The strong decrease in surface solar radiation due to aerosols produces strong surface radiative cooling. For oceanic layers of 10 m depth, and using Eq. (2) and a c_p of water equal to $\sim 4184\text{ J Kg}^{-1}\text{ K}^{-1}$, a value of 7 W m^{-2} for $\Delta F_{\text{surfnet}}$ translates to an oceanic water cooling rate of -0.015 K day^{-1} . For land, using a 1 m deep soil layer with a c_p value of $800\text{ J Kg}^{-1}\text{ K}^{-1}$, the estimated cooling rate is equal to -1.24 K day^{-1} for $\Delta F_{\text{surfnet}}$ equal to 15 W m^{-2} .

The drastic decrease of surface solar radiation due to aerosols, an effect which is called global dimming, is very important since it can modulate the hydrological cycle (e.g. Jacobson, 2001; Kaufman et al., 2002; Ramanathan et al., 2001). Recent studies (e.g. Lau et al., 2006) have shown that on time-scales of climate change, the cooling of the Earth's surface might lead to a gradual spin-down of the tropical water cycle and the eventual weakening of the Asian monsoons. The values of $\Delta F_{\text{surfnet}}$ are larger compared to those of ΔF_{TOA} and ΔF_{atmab} (Figs. 1 and 3, respectively), so that the aerosol DRE is much larger at the Earth's surface than at TOA or in the atmosphere. This is further discussed in the next section.

3.4 Ratio of top-of-atmosphere to surface shortwave aerosol direct radiative effect

The three types of DRE satisfy the conservation condition

$$\Delta F_{\text{TOA}} + \Delta F_{\text{surfnet}} + \Delta F_{\text{atmab}} = 0 \quad (3)$$

The atmospheric absorption of aerosols and $\Delta F_{\text{surfnet}}$ can not be directly estimated from satellite observations. When ΔF_{atmab} values are extremely small, then ΔF_{TOA} and $\Delta F_{\text{surfnet}}$ are essentially equal and opposite and this permits the surface DRE to be estimated from the DRE at TOA (Ramanathan et al. 2001), as in the case of purely

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scattering aerosols.

Computed values of the ratio $\Delta F_{\text{surfnet}}/\Delta F_{\text{TOA}}$ are given in Fig. 5. The ratio ranges from -10 to 10 over most of the oceanic and land areas, demonstrating the dominance of the surface to TOA aerosol DRE. Note that in cases of very small ΔF_{TOA} values, the ratio's magnitude can be much larger than 15 , but without any physical meaning, and hence such areas have been masked in Fig. 5 (white shaded areas e.g. in Sahara and Arabian peninsula). When the ratio takes values close to 1 , the magnitude of surface forcing equals that at TOA, which means that ΔF_{atmab} is negligible. In case of large negative $\Delta F_{\text{surfnet}}/\Delta F_{\text{TOA}}$ values, the ratio $\Delta F_{\text{atmab}}/\Delta F_{\text{TOA}}$ is also large when there are strongly absorbing aerosols in the atmosphere ($\omega_{\text{aer}} < 0.95$), e.g. over southern and eastern Asia, sub-Sahel, south Africa, and Europe in July, especially when surface albedo is large; this is the case of northern polar latitudes in July. Regions with reddish color correspond to identical sign values of DRE at TOA and surface; given that $\Delta F_{\text{surfnet}}$ is always negative (see Fig. 4), these regions must have negative ΔF_{TOA} values, i.e. they are characterized by planetary warming due to aerosols. This occurs over regions with large surface albedo and strongly absorbing aerosols above, e.g. Sahara, Arabian peninsula, Greenland.

Further, our results indicate that over many areas of the globe the surface DRE of aerosols can be up to 3 – 4 times larger than at TOA, in general agreement with the findings of Ramanathan et al. (2001) and Kaufman et al. (2002), but there are also many areas where the ratio exceeds 5 , as also reported recently by Chung et al. (2005).

3.5 Global and hemispherical averages of aerosol direct radiative effects

Global and hemispherical averages of SW DREs at TOA, in the atmosphere and at surface were also estimated, by including surface area weighting in the computations, and the results are given in Table 1 and Fig. 6. The largest DRE arising from natural plus anthropogenic aerosols is found at the surface, where it is about twice the DREs in the atmosphere and at TOA, irrespective of the hemisphere; the downward and absorbed

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fluxes are decreased by about 3.9 and 3.2 Wm^{-2} , respectively, on a global scale. In the atmosphere, solar radiation absorption increases by about 1.5 Wm^{-2} , and the globally averaged reflected solar flux to space increases by about 1.7 W m^{-2} . Our model value at TOA is in good agreement with other recent estimates from measurement-based approaches (e.g. Loeb and Manalo-Smith, 2005; Yu et al. 2006 review paper), ranging between -1.5 and -2.0 Wm^{-2} . For both ΔF_{atmab} and $\Delta F_{\text{surfnet}}$ there are large inter-hemispherical differences, with the largest effect of aerosols taking place in the more polluted and less cloudy northern hemisphere. More specifically, the northern hemispherical values for ΔF_{atmab} are larger than the southern ones by factors of 3.3 and 6.3 in January and July, respectively, while the corresponding factors for $\Delta F_{\text{surfnet}}$ are 1.7 and 2.5 , i.e. they are quite smaller. On the contrary, very small inter-hemispherical differences are found for ΔF_{TOA} , as a result of the combination of ΔF_{atmab} and $\Delta F_{\text{surfnet}}$. In general, the magnitude of global hemispherical aerosol DREs at surface and in the atmosphere is slightly (by factors 1.3 and 1.1 , respectively) larger in July than in January. Also, significant differences in the relative importance of the DREs at TOA, in the atmosphere and at the surface, are found between the two hemispheres. For example, the ratio $\Delta F_{\text{surfnet}}/\Delta F_{\text{atmab}}$ is about -1.7 in the northern hemisphere and -3.7 in the southern hemisphere. This inter-hemispherical contrast indicates the different nature of aerosols, being much more scattering in the cleaner southern hemisphere compared to the more absorbing nature of those in the northern hemisphere that includes many anthropogenic sources and most of the world's deserts. As for the spectral contribution to the total SW DREs (see Fig. 6), it appears that for all DRE components, both for summer and winter and both hemispheres, the largest contribution (about 70 – 80%) is in the UV-visible wavelengths.

A comparison of our computations of aerosol DREs at TOA and at the Earth's surface under all-sky conditions with other estimates based on satellite observations and multi-component aerosol models is presented in Table 2. Very few published results for $\Delta F_{\text{surfnet}}$ exist in literature, unlike for ΔF_{TOA} . The present estimate of all-sky global mean ΔF_{TOA} (-1.62 Wm^{-2}) is larger than previous estimates obtained with aerosol

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models (ranging from -0.24 to -1.28 Wm^{-2}), while it is very close to the estimates by Yu et al. (2004, 2006) based on measurement-based approaches. We note that estimates computed with coupled aerosol and general circulation models are not only based on a restricted number of spectral intervals but are also based on generated global distributions of cloud cover fractions that are not as accurate as those from satellite observations, as used in our study.

4 Summary and Conclusions

In this study, we quantified the global-scale direct radiative effect (DRE) of natural plus anthropogenic aerosols on solar radiation. This was accomplished by using a detailed spectral radiative transfer model covering the UV, visible, and near-IR wavelengths together with spectral aerosol optical properties (optical thickness, single scattering albedo and asymmetry parameter) taken from the Global Aerosol Data Set (GADS). This is in contrast to other similar studies often treating the whole solar spectrum as one spectral interval. The state of the atmosphere (clouds included) and that of the Earth's surface were specified using reanalysis data sets and satellite measurements. Therefore, the DRE on the outgoing SW radiation at the top of atmosphere (ΔF_{TOA}), on atmospheric absorption (ΔF_{atmab}), and on the downward and absorbed SW radiation at the Earth's surface (ΔF_{surf} and $\Delta F_{\text{surfnet}}$) were computed for realistic conditions of the Earth-atmosphere system for the 12-year period 1984–1995. The results are given for January and July at 1° latitude-longitude resolution for all-sky conditions, but also averaged for the two hemispheres and for the globe, and they represent a climatological assessment of aerosol DREs.

On a mean global basis, natural and anthropogenic aerosols together are found to increase the outgoing SW radiation at TOA (planetary cooling) by about 1.6 Wm^{-2} , and to radiatively heat the atmosphere (atmospheric heating) by increasing the absorption of solar radiation by about 1.6 Wm^{-2} . As a result, the downwelling and the absorbed SW radiation by the Earth's surface is found to be decreased (surface cooling) by 3.9

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and 3.2 Wm^{-2} , respectively, due to aerosols. Recently, Yu et al. (2006) presented in a review paper aerosol DRE values over land+ocean from measurement-based approaches under clear-sky conditions. These values when combined and adjusted to all-sky conditions are equal to -1.9 Wm^{-2} at TOA, and -3.4 Wm^{-2} at surface, i.e. they are very close to our computations (-1.6 and -3.2 Wm^{-2} , respectively). It should be noted that our computations refer to the DRE of natural plus anthropogenic aerosols. The effect of anthropogenic aerosols only, called direct climate forcing (DCF), can be derived nowadays from new-generation satellite instruments (e.g. MODIS), which are able to distinguish between fine (submicron) and coarse (supermicron) aerosols. Thus, recent estimates under all-sky conditions (e.g. Bellouin et al. 2005) give a DCF at TOA equal to $-0.8 \pm 0.1 \text{ W m}^{-2}$ and at surface equal to $-1.9 \pm 0.2 \text{ Wm}^{-2}$, values that correspond to about 50% of our computed total aerosol DRE. There are also other model-based estimates of DCF with smaller magnitude, such as those of -0.65 Wm^{-2} by Penner et al. (1998), -0.19 and -0.1 Wm^{-2} by Takemura et al. (2002) and (2005), respectively, and -0.35 Wm^{-2} by Chung et al. (2005).

The magnitude of the combined radiative effect of natural and anthropogenic aerosols at TOA (ΔF_{TOA}), which is equivalent to an increase in planetary albedo of the order of 1%, is very important for the climate of the Earth-atmosphere system, since it is comparable to that of climate forcing induced by elevated concentrations of greenhouse gases (about 2.4 Wm^{-2} , IPCC, 2001). Nevertheless, the largest DRE occurs at surface ($\Delta F_{\text{surfnet}}$, about double ΔF_{TOA}) having the potential for major effects on surface radiation budget (SRB), evaporation, and the hydrological cycle. The mean global enhanced absorption of solar radiation by 1.5 Wm^{-2} translates to an estimated atmospheric heating rate of 0.01 K day^{-1} . The major part of the computed DREs is for the northern hemisphere, which has most anthropogenic aerosol sources and desert areas, while larger DREs occur in July than in January.

According to our computations, a large percentage of the incident solar radiation at TOA is radiatively forced by aerosols. More specifically, these percentages can be as large as 13% at the Earth's surface and TOA, and 7% in the atmosphere. Furthermore,

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the aerosol effect on planetary albedo (ΔR_p) can even change sign. Thus, although it is found that in general, natural and anthropogenic aerosols together cool the Earth-atmosphere system, through scattering of SW radiation back to space, by up to about 15 Wm^{-2} , aerosols are also found to produce planetary warming (by up to 10 Wm^{-2}) over areas such as Sahara, Arabian peninsula, Siberia and Greenland. Regionally, the aerosol particles warm the atmosphere by absorbing solar radiation by up to 35 Wm^{-2} . The largest aerosol-induced atmospheric warming occurs above areas characterized by strongly absorbing mineral dust particles, especially over highly reflecting deserts. The estimated ΔF_{atmab} values translate into a tropospheric maximum heating rate of 0.98 K day^{-1} over land, and $<0.14 \text{ K day}^{-1}$ over oceans. At the Earth's surface, the downward and absorbed SW radiation is decreased by up to 45 and 30 Wm^{-2} , respectively, due to aerosol scattering and absorption. Regionally, the ratio of surface to TOA aerosol SW DRE changes from strong negative values of -10 to -15 to strong positive values up to 15, which indicates the important role of both natural and anthropogenic aerosols for surface processes and the surface radiation budget. This strong surface SW cooling combined with an associated atmospheric warming, can have important consequences on atmospheric circulation, since it can create more stable atmospheric conditions (by decreasing the vertical atmospheric temperature gradient) and lower surface evaporation, resulting in less clouds and precipitation, thus enhancing desertification processes, especially in semi-arid regions such as the Mediterranean basin or northern Africa.

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Table 1. Mean global and hemispherical (NH, SH) all-sky shortwave aerosol direct radiative effect (DRE, denoted as ΔF in W m^{-2}) for January and July. The DRE components are given in terms of: outgoing radiation at TOA (ΔF_{TOA}), radiation absorbed in the atmosphere (ΔF_{atmab}), downward radiation at surface (ΔF_{surf}), and net downward (absorbed) radiation at surface ($\Delta F_{\text{surfnet}}$).

		ΔF_{TOA}	ΔF_{atmab}	ΔF_{surf}	$\Delta F_{\text{surfnet}}$
January	Globe	1.72	1.41	−3.87	−3.13
	North Hemisphere	1.79	2.16	−4.96	−3.95
	South Hemisphere	1.66	0.65	−2.79	−2.31
	Globe	1.51	1.79	−4.0	−3.3
July	North Hemisphere	1.65	3.09	−5.83	−4.74
	South Hemisphere	1.37	0.49	−2.17	−1.86

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Table 2. Comparison of published shortwave aerosol DRE (in W m^{-2}) at TOA (ΔF_{TOA}^*) and at the Earth's surface ($\Delta F_{\text{surfnet}}$) under all-sky conditions.

	ΔF_{TOA}			$\Delta F_{\text{surfnet}}$		
	NH	SH	Global	NH	SH	Global
Present study	−1.72	−1.51	−1.62	−4.35	−2.09	−3.22
Yu et al. (2006)			−1.9			−3.4
Reddy et al. (2005)	−1.51	−0.58	−1.04			
Liao et al. (2004)			−0.93			
Yu et al. (2004) ^a	−2.08	−1.52	−1.8	−5.32	−2.56	−3.96
Yu et al. (2004) ^b	−1.52	−1.04	−1.28	−4.16	−1.84	−3.0
Takemura et al. (2002)	−0.17	−0.3	−0.24			
Jacobson (2001)			−1.23			

^a Obtained with MODIS+GOCART (Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport) integration

^b Obtained with GOCART alone

*In this Table, the sign of our ΔF_{TOA} (Present study) has been reversed with respect to that assumed throughout this study including Table 1, in accordance with the convention adopted by the other investigators.

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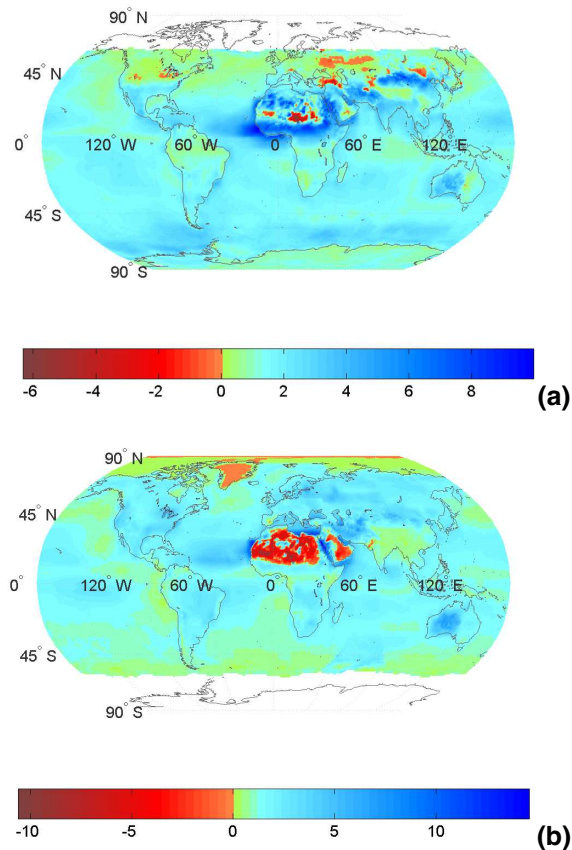


Fig. 1. Global distribution of the aerosol direct radiative effect (DRE) on the outgoing shortwave radiation at top-of-atmosphere (aerosol DRE ΔF_{TOA} , W m^{-2}), for **(a)** January and **(b)** July.

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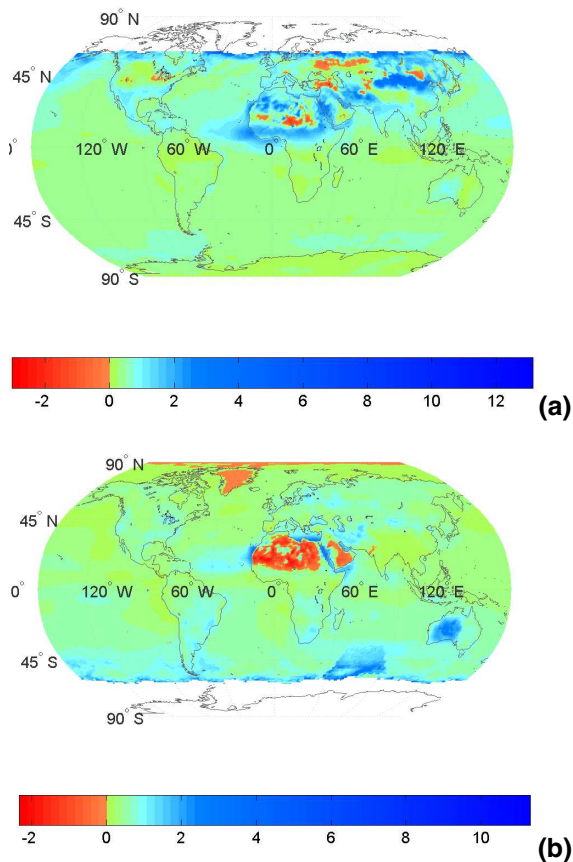


Fig. 2. Global distribution of change in planetary albedo (ΔR_p , in %) due to the aerosol direct radiative effect (DRE), for **(a)** January and **(b)** July.

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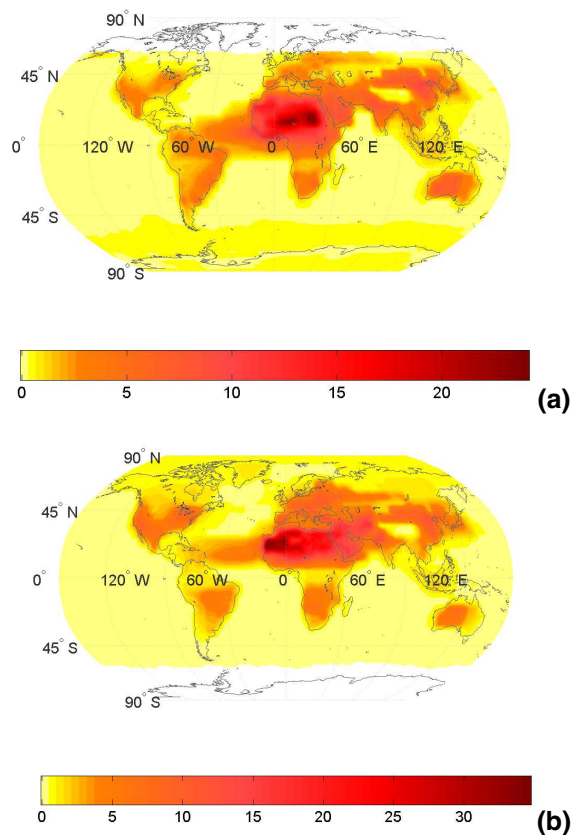


Fig. 3. Global distribution of the aerosol direct radiative effect (DRE) on the atmospheric absorption of shortwave radiation (aerosol DRE ΔF_{atmab} , W m^{-2}), for **(a)** January and **(b)** July.

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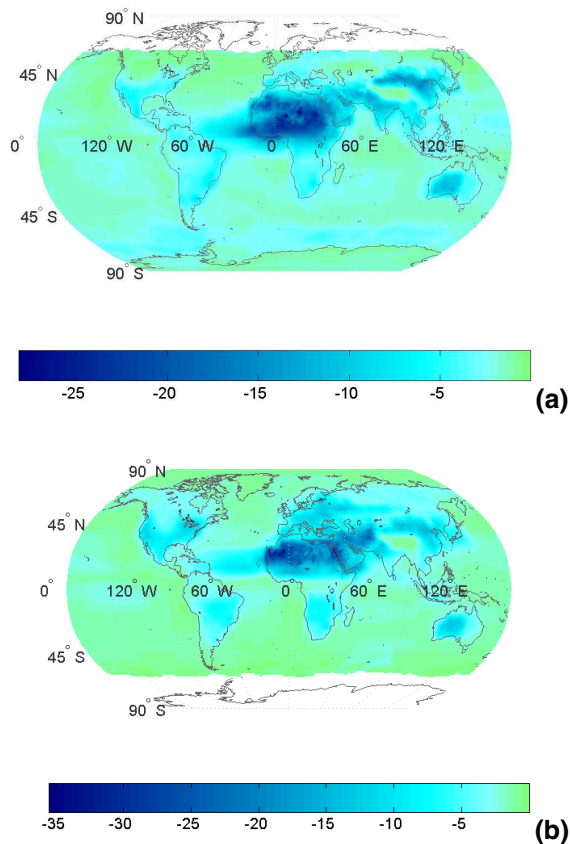


Fig. 4. Global distribution of the aerosol direct radiative effect (DRE) on the absorbed shortwave radiation by the Earth's surface (aerosol DRE $\Delta F_{\text{surfnet}}$, W m^{-2}), for **(a)** January and **(b)** July.

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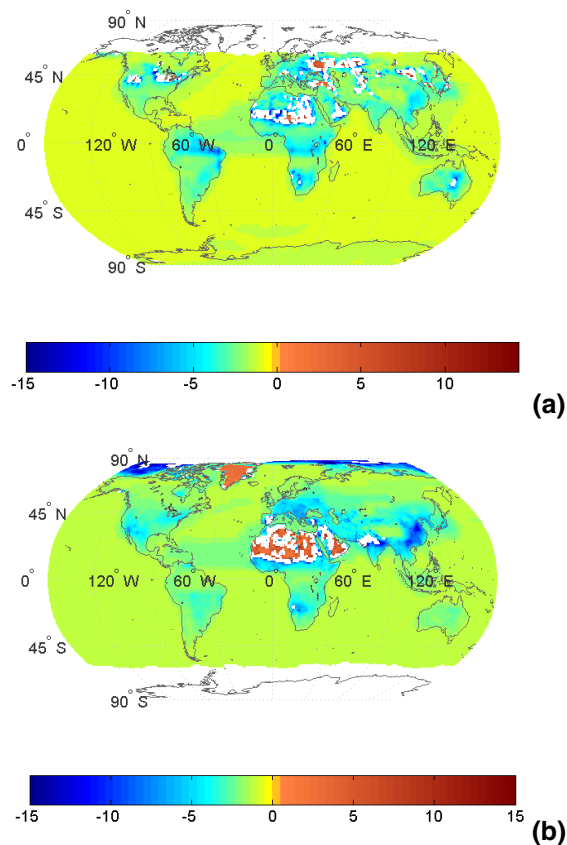


Fig. 5. Global distribution of the ratio of aerosol direct radiative effects $\Delta F_{\text{surfnet}}/\Delta F_{\text{TOA}}$, for **(a)** January and **(b)** July. Extremely small FTOA values, close to zero, which are associated with large ratios with no physical significance have been omitted.

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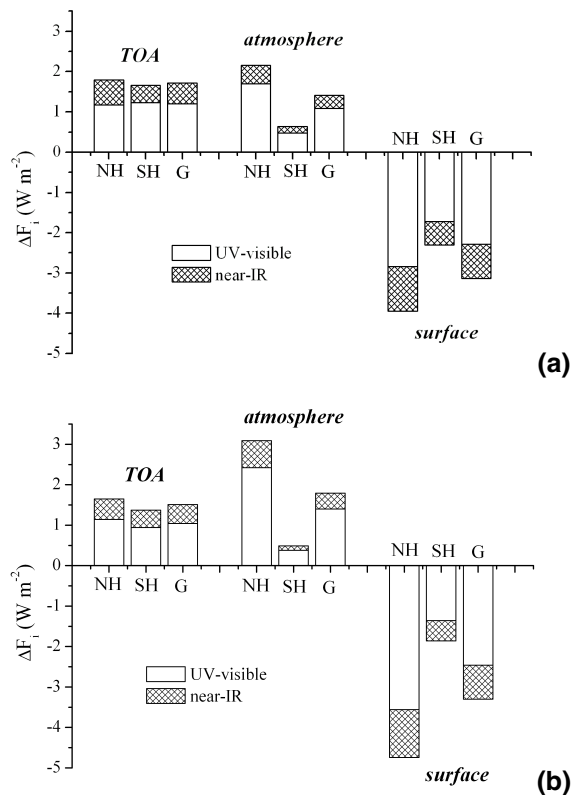


Fig. 6. Hemispherical and global averages of aerosol direct radiative effects (DRE) on the shortwave radiation at: TOA (ΔF_{TOA}), in the atmosphere (ΔF_{atmab}), and at surface ($\Delta F_{\text{surfnet}}$), for (a) January and (b) July. Each forcing is divided in ultraviolet (UV)-visible and near-infrared (IR) components. NH, SH and G correspond to North Hemisphere, South Hemisphere and Globe.